Magnetite in unequilibrated ordinary chondrites: evidence for an [17]O-rich reservoir in the solar nebula.

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Magnetite in unequilibrated chondrites is a key mineral for constraining low temperature processes in the solar nebula and in the parent asteroids of chondrites. A small fraction of the unequilibrated ordinary chondrites (UOC) have magnetite which is typically associated with carbides, sulfides, and Fe-Ni metal [1,2,3]. The assemblages were interpreted as the products of reaction of metal with S, C, and O in the solar nebula [1], or the results of oxidation processes in the parent asteroid, possibly by H_2O-rich vapor [3]. It was also suggested that there were two-magnetite-forming stages, pre-accretion and post-accretion [2]. To better understand the formation of the magnetite, and low temperature processes in the solar nebula and in the parent asteroids of the chondrites, we have performed *in situ* measurements of O-isotopic compositions of magnetite in the Semarkona (LL3.0) and Ngawi (LL3.2/3.7) ordinary chondrites. Saxton et al. [4] reported data from four points on a Semarkona magnetite grain that yielded a mean $\Delta^{17}O$ ($=\delta^{17}O - 0.52 \cdot \delta^{18}O$) of ~ 3‰. Our data (20 points on 12 grains) cluster near $\Delta^{17}O$ = 5.3‰, the highest value in a material that originated in the solar system.

We investigated three Semarkona thin sections (UCLA 229, USNM 1805-5, and UNM 312) and one Ngawi thin section (USNM 2483). These were characterized using a petrographic microscope and back-scattered electron imaging and 10 Semarkona and 2 Ngawi magnetites were chosen for O-isotopic study. Among the 10 Semarkona magnetites, two are in a low-FeO porphyritic olivine chondrule, two in a porphyritic pyroxene chondrule, and the other six in the matrix. The two Ngawi magnetites are sited in the matrix. Measurements on the Cameca 1270 were carried out during July-November 1996. A defocused Cs^+ beam was used to sputter areas having diameters ~ 12 to 25 μm. Negative ^{16}O ions were measured by a Faraday cup, and other isotopes collected on an electron multiplier. The maximum OH^- ion interference to the $^{17}O^-$ signal was 0.5‰, but typically less than 0.1‰. More detailed analytical procedures are found in Choi et al. [5].

Our results on the 10 Semarkona and 2 Ngawi magnetite grains are plotted in Fig. 1. The magnetite has a mean $\Delta^{17}O$ of ~ 5.3‰ much higher than the bulk silicates ($\Delta^{17}O$ of ~1.1‰ [6]). Only interstellar Al_2O_3 grains have compositions more ^{17}O-rich [7,8] than these magnetite of undisputed solar system origin. The magnetite O-isotopic data fall near a mass fractionation line (the slope of a regression line is 0.63 rather than 0.52). There is no difference in O-isotopic compositions of the magnetites in chondrules and matrix. Semarkona and Ngawi magnetites fall on the same line, implying they formed by the same oxidant. The magnetite data show a large range of about 13‰ in $\delta^{18}O$, suggesting a Rayleigh-type fractionation involving a limited supply of oxidant, which implies an asteroidal setting for the magnetite formation.

Because there was no oxygen in the precursors of the magnetite (Fe or FeS [1,2,3]), the O-isotopic composition of the magnetite reflects that of the oxidant. If the magnetite formed by a limited supply of oxidant whose oxygen was completely consumed by forming magnetite, the mean O-isotopic composition of magnetite is the same as the initial composition of the oxidant; $\delta^{18}O$ = 0.9‰ for our data. Another estimate of the initial composition of the oxidant is possible by assuming that the lightest O-isotopic composition (-4.0‰ in $\delta^{18}O$) represent the first magnetite formed. In an asteroidal setting, the most likely oxidant for magnetite formation is H_2O [3]. The maximum fractionation between magnetite and liquid H_2O is -13.6‰ at 390 K [9]. In the UOC parent asteroid, H_2O probably existed as a gaseous phase when magnetite formed. The maximum fractionation between magnetite and gaseous H_2O is -10.5‰ at 500 K [10]. If the Semarkona magnetite formed at this condition, the initial $\delta^{18}O$ of the H_2O should be less than 6.5‰. If the lightest O-isotopes we found is, for example, the average of first 20 wt.% magnetite formed during the Rayleigh-type fractionation, the initial O-isotopic compositions of the H_2O is calculated to 5.5‰ in $\delta^{18}O$.

A key question is whether the H_2O oxidant of the magnetite originated from same major nebula

gaseous reservoir that contributed [17]O-rich oxygen to the UOC silicates. If so, this gas had $\Delta^{17}O \geq 5.3$‰; the refractory solid reservoir should have had $\Delta^{17}O \leq 0$‰.   The O-isotopic compositions of the bulk UOC silicates [6] fall along a slope 0.7 mixing line; those of UOC chondrule [6] a slope 0.82.   If we take the average of these mixing lines, it meets with a fractionation line having $\Delta^{17}O = 5.3$‰ at $\delta^{18}O = 22$‰, similar to some estimates of the nebular gas [11,12].   The O-isotopic composition of the gaseous reservoir which formed the UOC silicates is then $\delta^{18}O = 22$‰ $+ \Delta^{18}O$, where $\Delta^{18}O$ is the equilibrium fractionation factor between gaseous oxygen and silicates. The model requires a large negative fractionation perhaps about 15-23‰ from the high temperature nebula gas to the asteroidal H_2O. An alternative possibility for the origin of the H_2O is that it represents a foreign component (e.g., originating in the outer solar system).   Some refractory oxides of interstellar origin are heavily enriched in [17]O [7,8].   The

addtion of a tiny fraction of such extreme oxygen into the common UOC oxygen could produce a reservoir having the O-isotopic compositions we observe in UOC magnetite.

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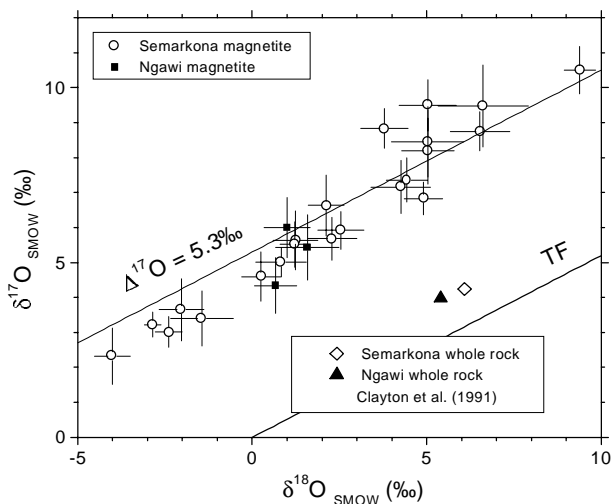

Fig. 1. Oxygen isotope compositions of points on Semarkona and Ngawi magnetite grains. One sigma error bars are shown. Data for bulk chondrites [6] are shown as references.   The compositions of magnetite are clearly distinguished by larger $\Delta^{17}O$ (average = 5.3‰) values from those of bulk chondrites.   Semarkona magnetite shows wide range of $\delta^{18}O$ along the fractionation line (the slope of the regression line for the Semarkona magnetite is 0.63 rather than 0.52); three data from the Ngawi magnetite fall in a small O-isotopic composition field.